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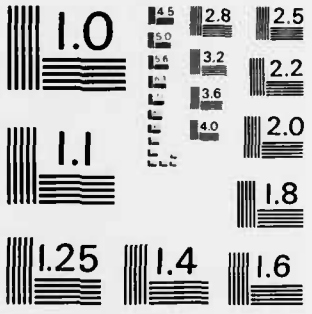

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by

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THEORETICAL ASPECTS OF LASER-INDUCED PERIODIC SURFACE STRUCTURE FORMATION

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ABSTRACT

Laser-induced periodic pattern formation has been observed on a variety of substances. In particular, low-power lasers have been used to deposit a pattern on a metal surface. For a relatively smooth surface grating, this pattern can be explained in terms of a perturbative solution of Maxwell's equations. However, as the surface grating is enhanced by this initial deposition, the perturbation solution breaks down. An alternate non-perturbative solution of Maxwell's equations for such rough surfaces is considered here. Moreover, other possible mechanisms that may assist pattern formation are discussed, such as field-enhanced evaporation and surface migration.

INTRODUCTION

Over the past several years, a number of researchers [1-17] have observed periodic patterns developing on the surfaces of solids that were exposed to laser radiation. These patterns have been seen on various metals, insulators, and both doped and pure semiconductors. Most of these experiments were carried out with Nd:YAG or ND:glass lasers tuned to wavelengths between 1.0 μm and 10.0 μm. To obtain these patterns, laser power densities of between 10 MW/cm^2 and 1 GW/cm^2 were employed. Above this range, surface melting would occur, while below this level, no discernible surface pattern was observed.

This laser-induced periodic surface structure has several characteristics. First, the surface pattern consists of a number of parallel grooves running perpendicular to the $\vec{E}$ field of the incident laser. Second, Oron and Sorensen [8] demonstrated that the underlying surface lattice has no effect on the shape of the laser-induced surface structure. On the other hand, Isenor [6] has shown that the existence of scratches on the surface can have a significant effect on the final surface structure. In fact, if the surface is randomly scratched, no periodic surface structure will form. Both Brueck and Ehrlich [12] and van Driel and his colleagues [13] have also observed the development of a weaker secondary pattern parallel to the incident $\vec{E}$ field. With this secondary structure, the surface takes on a scalloped appearance. Finally, these surface patterns produce large drops in the reflectivity of several metals [3-4].

Most of the work so far has considered laser-induced periodic structures on surfaces that already had a certain composition. Needless to say, this necessitates large laser power densities (greater than 10 MW/cm^2) in order to provide sufficient energy for the surface atoms to rearrange. However, Brueck and Ehrlich [12] showed that only a modest amount of laser power (less than 10 W/cm^2) is needed to establish the periodic surface structure when the constituent atoms are deposited from the gas phase. In particular, they showed that Cd, Zn and Al in organometallic compounds could be photochemically deposited on Si or SiO_2 substrates in ordered arrays.

To understand these observations and resolve any difficulties, one must consider how this laser-induced periodic surface structure is produced. There have been three attempts to explain these effects. First, since many metals have a plasma frequency around the frequency of the incident laser radiation, surface plasmons will be excited [11-12]. These plasmons will couple with the electromagnetic field to form a surface polariton. Since this polariton produces maxima and minima in the $\vec{E}$ field of the surface, the surface atoms will rearrange themselves to minimize their energy in this $\vec{E}$ field. Second, Maracas et al [7] have observed that the periodic surface structure can be considered a standing wave. This wave would have a velocity very close to that of a longitudinal acoustic phonon of the substrate. Thus, the surface pattern may well be a phonon excited by the laser that was "frozen" in place by the cooling of the lattice. Finally, van Driel and his associates [15] have proposed an extensive theory based on the small inhomogeneities that exist in the surface layer. This initial surface roughness will interact with the incident laser beam to produce a dipole moment in the surface layer (a "radiation remnant"). The field generated by this dipole layer can interfere with the refracted beam in the substrate below the surface. This interference will lead to inhomogeneous energy absorption and thus the redistribution of the surface atoms.

In the following section, the theory of the surface enhanced $\vec{E}$ field produced by relatively smooth surfaces and the resultant pattern formation will be viewed. The inapplicability of this approach to rough gratings will be demonstrated. To overcome this limitation, a non-perturbative solution of Maxwell's equations will be discussed. Finally, our results will be presented along with suggested improvements.

THEORY OF SMOOTH GRATINGS

Incident radiation converted to surface plasmon excitations via surface roughness has been investigated extensively in the past decade [18]. The classical method of Rayleigh [19] has often been used to solve the problem of weak scattering [20]. Brueck and Ehrlich [12] also adapted this formalism to describe the phenomena of laser-induced pattern formation. Here, we will briefly outline the theory of Brueck and Ehrlich and discuss its validation and limitation.

Let a light wave of frequency ω represented by an electric field

$$\vec{E}_{inc}(\vec{r},t) = E_i e^{-ik_o z} e^{-i\omega t} \tag{1}$$

be incident on the metal surface at a normal angle, on the xz-plane. If we express the surface roughness in terms of a Fourier expansion,

$$z = \xi(x,y) = \sum_{\vec{g}} u_g e^{i\vec{g}\cdot\vec{r}_{\parallel}} \quad , \vec{g} = (g_x, g_y), \vec{r}_{\parallel} = (x,y), \tag{2}$$

and assume that each amplitude u_g is small compared to the wavelength of the incident light, then taking the dielectric function as a step function,

$$\varepsilon(\omega,\vec{r}) = \varepsilon(\omega)\Theta(z-\xi) + \Theta(\xi - z), \tag{3}$$

we can write down the solutions of the Maxwell's equations separately for the two media. In particular, for $g^2 > \omega^2/c^2$ and $z > \xi$, we have

$$\vec{E} = \vec{E}_i e^{-ik_o z} + \vec{E}_r e^{ik_o z} + \vec{E}_{sp} e^{i\vec{g}\cdot\vec{r}_{\parallel}} e^{-k_g z}, \tag{4}$$

where $k_g^2 = g^2 - \omega^2/c^2$. By imposing the boundary conditions, i.e., the local

tangential components of the electric and magnetic fields have to be continuous across the actual surface $z = \xi(x,y)$, we can express the reflected and surface plasmon fields, $\vec{E}_r$ and $\vec{E}_{sp}$ respectively, in terms of $\vec{E}_i$ by writing

$$e^{ik_o\xi} \approx (1 + ik_o\xi) \tag{5}$$

and assuming $g\xi << 1$ in the continuity equations. If we consider only the p-wave scattering, i.e., the light polarized in the x-direction, the expression of Brueck and Ehrlich for $\vec{E}_{sp}$ can then be easily obtained. If we assume a linear relationship between the film growth rate and the local intensity,

$$\frac{dT}{dt} \propto I(x) = |\vec{E}\cdot\vec{E}|, \tag{6}$$

then the suggested equation of the grating depth u_p is obtained by assuming a spatial profile of $T(x) = T_o + u\cos(gx)$ and neglecting the $|\vec{E}_{sp}|^2$ term.

The foregoing theory was developed under the assumption that the surface roughness is small, $g\xi << 1$. However, as more metal is deposited, the roughness increases. In the perturbative theory of Brueck and Ehrlich, $\vec{E}_{sp}$ is proportional to the roughness u. Therefore, the first consequence of increased roughness is that $|\vec{E}_{sp}|^2$ can no longer be neglected. This point may already be reached when the grating is only a few layers deep. Moreover, the damping of $\vec{E}_{sp}$ can no longer be measured from the mean surface. An increase in roughness can lead to sufficient enhancement of $\vec{E}_{sp}$ that localized evaporation of metal atoms could play an important role. Transfer of large amounts of energy to the metal-carbon bond, such as in the deposition of Cd on Si due to the dissociation of gaseous $Cd(CH_3)_2$, may also induce sufficient translational motion along the surface so that adsorption does not occur at the dissociation site. Such dissociation under several possibly physisorbed organometallic layers could lead to trapping of the organic radical in the vicinity of the surface. This "cage" effect could lead to the reformation of the organic metal bond. Finally, the effect of surface roughness on the dielectric function might be important. Such increases of surface area could substantially alter the magnitude of this function, which in turn could enhance the rate of pattern formation.

GENERALIZATION TO ROUGH GRATINGS

The foregoing considerations lead us to conclude that an explanation of deep pattern formation requires a non-perturbative treatment of plasmon formation, and at the present time a numerical solution of Maxwell's equations seems inevitable. Recently [21] there has appeared a solution of the problem of a square-well metallic grating in an applied electromagnetic field (FIG. 1b). Such a solution is restricted to explaining the most qualitative features of sinusoidal grating formation. We have established a general formulation of the problem which is capable in principle of handling gratings of any shape or depth.

In the H polarization, Maxwell's equations can be written as [22]

$$\frac{\partial^2 H_y}{\partial z^2} + \frac{\partial^2 H_y}{\partial x^2} + \varepsilon(x)k_o^2 H_y = \frac{\partial H_y}{\partial x}\,\frac{\partial}{\partial x}\,[\ell n\varepsilon(x)], \tag{7}$$

where H_y is the component of the magnetic field H in the direction perpendicular to the lattice vector of the grating (FIG. 1). For square-well gratings, H_y is separable,

$$H_y(x,y) = Z(z)\,X(x)\,, \tag{8}$$

and Eq. (7) can be written as two coupled first-order differential equations:

$$\frac{\partial Z^2}{\partial z^2} = \Lambda^2 Z \tag{9a}$$

$$\frac{\partial^2 X}{\partial x^2} - \frac{\partial}{\partial x}\left[\ell n\,\varepsilon(x)\right]\frac{\partial X}{\partial x} + \left[\varepsilon(x)k_o^{\,2} - \Lambda^2\right]X = 0. \tag{9b}$$

When the applied radiation is incident normal to the grating, it is found that Λ is the solution of the equation

$$1 - \cos(\beta d/2)\cos(\alpha d/2) + 1/2(\varepsilon\alpha/\beta + \beta/(\alpha\varepsilon))\sin(\beta d/2)\,\sin(\alpha d/2) = 0\,, \tag{10}$$

where d is th period of the grating, ε is the dielectric constant of the grating, $\alpha = (k_o^{\,2} - \Lambda^2)^{1/2}$ and $\beta = (\varepsilon k_o^{\,2} - \Lambda^2)^{1/2}$.

The problem to be considered (FIG. 1c) is that of a multilayered grating. In each layer the grating is periodic and the general solution to Eq. (9) for the n-th layer may be written as

$$\psi_n^{II} = \sum_{\ell} X_{n\ell}(x)\left[A_{n\ell}\,e^{i\Lambda_{n\ell}z} + B_{n\ell}\,e^{-i\Lambda_{n\ell}z}\right], \tag{11}$$

where the coefficients A and B are to be determined by the boundary conditions between layers, and $\Lambda_{n\ell}$ is the ℓ-th solution to Eq. (10) for the n-th layer. By applying the boundary condition of continuity of H_y and $\frac{\partial}{\partial z}$ between layers, we can establish the recursion relations for A and B,

$$\vec{A}_n = E_{n,n-1}\vec{A}_{n-1} + F_{n,n-1}\vec{B}_{n-1} \tag{12a}$$

$$\vec{B}_n = C_{n,n-1}\vec{A}_{n-1} + D_{n,n-1}\vec{B}_{n-1}\,, \tag{12b}$$

where the matrices C, D, E, F are determined by the periodic structure of each layer. Furthermore, we can establish boundary conditions between the top two layers and the two lowest layers. In the topmost (infinite) layer, the eigensolution is written as [21]

$$\psi^I = e^{-ik_o z} + \sum_{n=-\infty}^{\infty} R_n\, e^{ik_o[\gamma_n x + (1-\gamma_n^{\,2})^{1/2} z]}. \tag{13}$$

In the lowest (infinite layer) we have

$$\psi^{III} = \sum_{n=-\infty}^{\infty} T_n\, e^{ik_o[\gamma_n x - (\varepsilon - \gamma_n^{\,2})^{1/2} z]} \tag{14}$$

where $\gamma_n = \frac{n\lambda}{d}$.

It is now possible to establish

$$\vec{A}_1 = a\vec{D} + b\vec{R} \tag{15a}$$

$$\vec{B}_1 = b\vec{D} + a\vec{R}, \tag{15b}$$

where $\vec{D}$ is a unit vector. In addition,

$$\vec{A}_N = L\vec{B}_N, \tag{16}$$

where N labels the penultimate (finite) layer, and the matrix L is determined by the parameters which characterize that layer. However, by applying the recursion relations of Eq. (12), it is possible to establish

$$\vec{A}_N = J_1\vec{D} + J_2\vec{R} \tag{17a}$$

$$\vec{B}_N = K_1\vec{D} + K_2\vec{R} \tag{17b}$$

Rearranging (16) and (17) gives

$$\vec{R} = (J_2 - LK_2)^{-1}(LK_1 - J_1)\vec{D}, \tag{18}$$

and by employing Eq. (12) once more we arrive finally at a matrix equation for the vectors $\vec{A}_M$ and $\vec{B}_M$,

$$\begin{pmatrix}\vec{A}_M\\ \vec{B}_M\end{pmatrix} = \prod_{n=2}^{M}\begin{pmatrix}E_{n,n-1} & F_{n,n-1}\\ C_{n,n-1} & D_{n,n-1}\end{pmatrix}\begin{pmatrix}a & b\\ b & a\end{pmatrix}\begin{pmatrix}\vec{D}\\ \vec{R}\end{pmatrix}. \tag{19}$$

RESULTS AND DISCUSSION

We have made a preliminary calculation for a square-well silver grating. The field intensity $|\vec{E}(z,x)|^2$ is averaged over x at each of two values of z (z = 0 and z = -h), and the ratio $R = \overline{|E(z=-h)|^2}/\overline{|E(z=0)|^2}$ is plotted as a function of the well-depth (roughness parameter) h (FIG. 2). R is thus the ratio of intensity in the "peak" region of the grating to that in the "well" region. For very shallow gratings (h < 0.5 nm), or about two monolayers) it is seen that $\overline{|E(z=0)|^2} > \overline{|E(z=-h)|^2}$, which supports the perturbation description of Brueck and Ehrlich. This would explain how a particular grating establishes itself out of the "noise" of microscopic roughness. It also explains why the establishment of such a grating for low-power lasers requires the photodissociation of an organometallic compound and does not occur in the presence of metal vapor alone. However, at a certain depth (around 0.5 nm) $\overline{|E(z=-h)|^2}$ begins to increase very rapidly. It seems here that the grating structure will only continue to reinforce itself by a mechanism other than photodeposition. Such a mechanism might be photoevaporation of the grating itself, whereby the trough regions are "excavated" by the field. Finally, we note that as $h \to \infty$, $\overline{|E(z=-h)|^2} \to 0$. This is consistent with an upper unit to the growth of the grating. Work is currently underway to determine whether these qualitative trends are consistent also with sinusoidal gratings.

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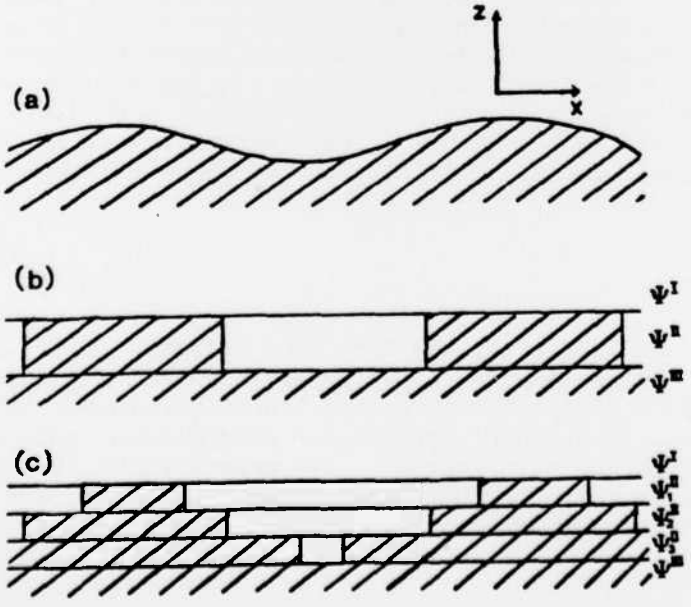

FIG. 1.

(a) Sinusoidal grating. The hatched area represents the metal.
(b) Square-well grating showing a separation into three layers, one of which is periodic in the x-direction, and two of which are uniform.
(c) Generalization of the square-well grating in which there are three periodic layers.

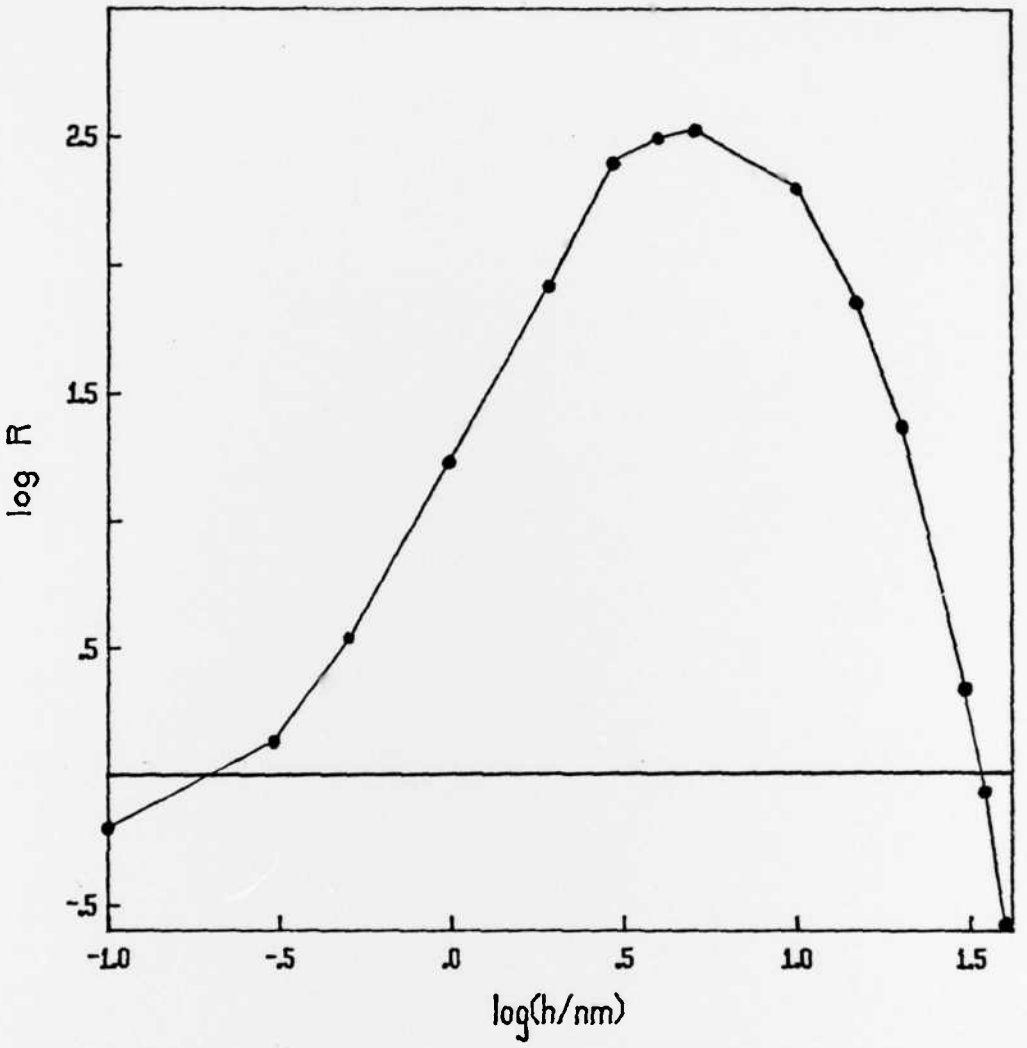

FIG. 2. Log-log plot of the ratio $R = \overline{|E(z=-h)|^2}/\overline{|E(z=0)|^2}$ against the grating depth h, for a silver grating with d = 1050 nm and λ = 700 nm. The horizontal line corresponds to R = 1. Above the line the intensity is greatest in the region of the well (z = -h); below the line the intensity is greatest in the region of the peaks (z = 0) of the grating. Clearly the latter condition pertains for very smooth gratings ($h \to 0$) and also for very rough gratings ($h \to \infty$)

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